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KRYPTON 85
A REVIEW of the LITERATURE
and
an ANALYSIS of
RADIATION HAZARDS

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The in vivo internal behavior of ^{85}Kr is discussed in detail in appendix A and preliminary desaturation curves obtained with rats are presented. The review includes 280 references.

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A REVIEW of the LITERATURE and an ANALYSIS of RADIATION HAZARDS

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JAN 1972

ENVIRONMENTAL PROTECTION AGENCY
Office of Research and Monitoring
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FOREWORD

Krypton-85 is a long-lived, fission-product, noble gas which is released to the atmosphere in large quantities by the nuclear industry, primarily by reactor fuel reprocessing plants. Although development of the technology needed to collect the krypton-85 at reprocessing facilities is nearing fruition, the atmospheric build-up of krypton-85 is expected to continue for some time due to the rapid growth of the nuclear power industry. The present atmospheric inventory of about 60 megacuries is more than twice the inventory of a decade ago and is increasing rapidly. The distribution of ^{85}Kr is essentially global once it is released, with radioactive decay ($T_{1/2}$ = 10.76 yr) being the only important removal mechanism.

The current permissible ^{85}Kr concentration values are based on calculations and extrapolations rather than on the results of direct experimental investigation of the effects of ^{85}Kr on living animals. Thorough investigation of the physiological behavior and effects of ^{85}Kr in living animals is, therefore, imperative. This report summarizes the background information upon which studies of this type, being undertaken at the Eastern Environmental Radiation Laboratory, Montgomery, Alabama, are based. Additional information is sought on a continuing basis, and the interest and comments of all those concerned with radiological and environmental health are solicited.

William A. Mills

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ABSTRACT

This review summarizes most of the existing information on ^{85}Kr. Major subject areas covered are (1) physical, chemical and radiological data, (2) maximum permissible concentration in air $(MPC)_a$ and its rationale, (3) source data, (4) atmospheric concentrations and dose estimates near reprocessing facilities and worldwide, (5) proposed control methods, (6) uses in science, especially medicine, and industry, (7) calculations of dose to various organs and their relationship to the $(MPC)_a$, (8) unexplained noble gas phenomena, and (9) methods of sampling and analysis.

The *in vivo* internal behavior of ^{85}Kr is discussed in detail in appendix A and preliminary desaturation curves obtained with rats are presented. The review includes 280 references.

ACKNOWLEDGMENTS

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INTRODUCTION

Krypton 85 is one of the most important gaseous contaminants produced in nuclear fission. Public health concern has centered on its release to the atmosphere during reactor operations and especially during fuel reprocessing. Rapid expansion of the medical, scientific and industrial uses of ^{85}Kr have made it of practical importance to the health physicist. The literature on ^{85}Kr is singularly parochial and is spread through many disciplines with little cross-referencing. This report reviews much of this literature and provides general information and references regarding ^{85}Kr. The reference list is not complete, particularly with respect to some government reports from this country and abroad which are difficult to locate and obtain, and to the rapidly proliferating field of medical uses. This report specifically attempts to:

1. Furnish physical, chemical, and radiological data on ^{85}Kr
2. Review sources, yields, and amounts released in different operations
3. Review the current maximum permissible concentrations in air $(MPC)_a$ values and their rationale
4. Review the status of ^{85}Kr as an environmental contaminant and proposed methods of control
5. Enumerate a number of uses for ^{85}Kr in science, especially medicine and industry
6. Evaluate the radiation hazard associated with ^{85}Kr and relate it to existing limits
7. Review methods that have been successfully used to collect, prepare and analyze ^{85}Kr.

BACKGROUND INFORMATION

CHARACTERISTICS

CHEMICAL

The family of noble gases that includes krypton has been traditionally regarded as chemically inert. Modern studies have revealed, however, that the more polarizable members can participate in ionic or covalent bonding, under appropriate conditions, with highly reactive elements such as fluorine and oxygen (1,2) and that clathrates[1] can be formed with water

[1] A clathrate is a solid that incorporates a gas into voids in its crystal structure. See figure 1.

(hydrates) and a number of organic solvents (1,3,4 5). Most of these compounds dissociate at physiological temperatures but some of the organic clathrates, including hydroquinones, are relatively stable at normal temperatures (4,5). The structure of hydroquinone clathrate is shown in figure 1. Several authors have postulated clathrate formation involving side chains on body proteins to explain the observed narcotic effects of xenon, the most reactive noble gas, and other gases that produce similar reactions (3,6,7,8). The noble gases are highly soluble in non-polar solvents, including body lipids, with solubility decreasing in order of radon, xenon, krypton, argon, neon, and helium (9). Solubility is discussed in more detail in Appendix A.

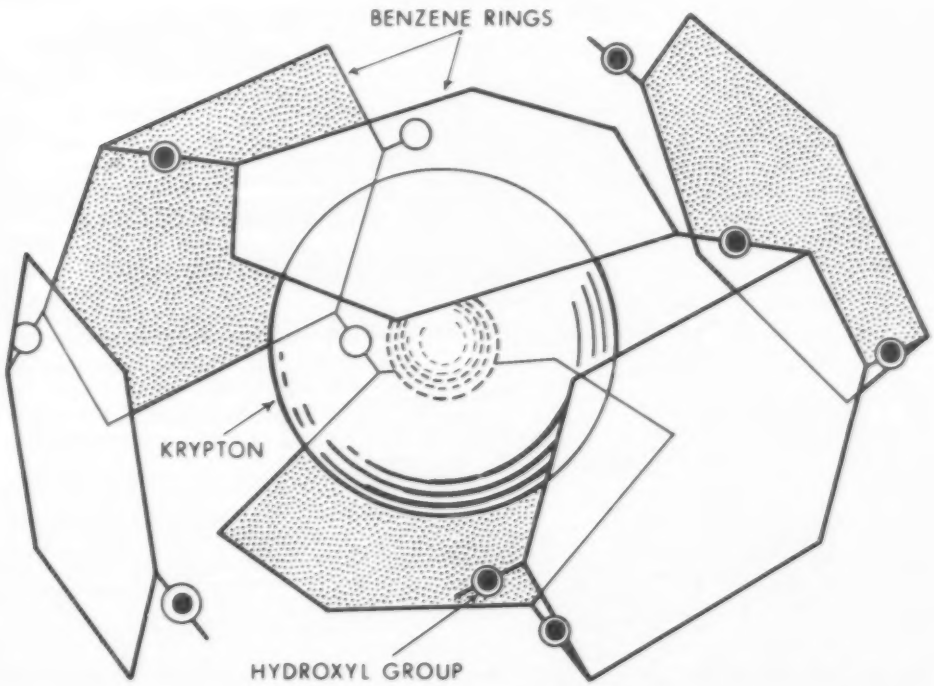

Figure 1. Scheme of hydroquinone clathrate, after Balek (5).

PHYSICAL DATA

Cryogenic Reference Data (10) and the *Radiological Health Handbook* (11) give the following information concerning krypton:

Atomic weight (naturally occurring) = 83.9
Melting point = -157.2° C (-250.9° F)
Boiling point = -153.3° C (-244° F)
Triple point = -157.2° C, 548.2 mm Hg (-251° F and 10.6 PSIA)
Critical point = -63.8° C (-82.8° F) and 41,165 mm Hg (796 PSIA)

One kg of natural krypton occupies a volume of 287.45 liters at NTP (20° C, 760 mm Hg) or 266.79 liters at STP (0°C 760 mm Hg).

The naturally occurring isotopes of krypton and their percentages of natural abundance are ^{78}Kr (0.35%), ^{80}Kr (2.27%), ^{82}Kr (11.56%), ^{83}Kr (11.55%), ^{84}Kr (56.9%) and ^{86}Kr (17.37%). Radioactive isotopes of krypton include 74-77, 79, 79m, 81, 81m, 83m, 85, 85m, 87-95, and 97.

RADIOLOGICAL DATA

The following data are given by the *Radiological Health Handbook* (11) or the *National Bureau of Standards* (12) for ^{85}Kr:

Half life = 10.76 years
Emissions: Beta; E_{max} = 0.672 MeV, $\bar{E}$ = 0.249 MeV, frequency = 99.59% (11) or 99.56% (12). A 0.16 MeV E_{max} beta, which is usually ignored in calculations, is associated with the 0.514 MeV gamma.
Gamma; E = 0.514 MeV, Branching ratio = 0.41% (11) or 0.443% (12).

Revisions in the published values have occurred and are the cause of most differences in doses or dose rates calculated by different authors.

SOURCE INFORMATION

The amount of ^{85}Kr formed depends on a number of factors including the specific heavy nuclei being fissioned, the neutron flux, the neutron energy spectrum, and the irradiation time. The amount present at analysis will be determined by the foregoing factors and the time that has elapsed between irradiaton and analysis, or cooling time. The general expression for the amount of ^{85}Kr present in a reactor or fuel element is:

$$C = 8.4 \times 10^5 P_i Y_i (1 - e^{-\lambda T}) e^{-\lambda t} \qquad (13)$$

where: C = curies of ^{85}Kr present
P_i = total nuclear power supplied by reactor system i, MW
Y_i = fission yield of system for ^{85}Kr
λ = decay constant for ^{85}Kr = 1.76×10^{-4} days^{-1}
T = irradiation time (days)
t = cooling time (days)

Various estimates of Y_i in the literature include:

Y_i (^{235}U, thermal neutrons) = 0.00293 (14); 0.00273 (15); 0.00306 (16)
Y_i (^{235}U, fission neutrons) = 0.00310 (16)
Y_i (^{239}Pu, thermal neutrons) = 0.00099 (15); 0.0012 (16)
Y_i (^{239}Pu, fission neutrons) = 0.001446 (16)
Y_i (^{239}Pu, fast neutrons) = 0.00076 (17)
Y_i (^{233}U, thermal neutrons) = 0.0058 (14)

An estimate of average ^{85}Kr production is 0.2 kCi per megawatt of energy produced (1 year operation, 1 day cooling)(18).

PRESENT $(MPC)_a$ VALUES AND RATIONALE

The presently accepted maximum permissible concentrations in air for ^{85}Kr, as established by the AEC (19), NCRP (20), and ICRP (21), are 3 x 10^{-7} $\mu Ci/cm^3$ (3 x 10^5 pCi/m^3) for unrestricted areas, 10^{-5} $\mu Ci/cm^3$ (10^7 pCi/m^3) for occupational exposure for 40 hours weekly, and 3 x 10-6 $\mu Ci/cm^3$ (3 x 10^6 pCi/m^3) for occupational exposure for a 168-hour week.

The $(MPC)_a$ values and the estimated doses in most published projections are based on calculations of the external dose received by a person standing in an infinite hemispheric cloud of the radioactive gas. This is standard procedure for noble gases.[2] Internal absorption or concentration is not considered. The occupational $(MPC)_a$ for a 40-hour week is calculated from the formula:

$$(MPC)_a = \frac{0.024\ R}{\Sigma(E)}\ \rho_a\ (\rho_a/\rho_t)\ \mu Ci/cm^3$$

where: R = dose permitted in one week (rem)
ρ_a = density of air (0.00129 g/cm^3)
ρ_a/ρ_t = stopping power of air relative to tissue (= 1/1.13 for beta and secondary electrons produced by x or γ radiation)
$\Sigma(E)$ = effective energy per disintegration (MeV)

Two sub-categories are recognized. If the radiation emitted is gamma radiation or beta radiation with maximum energy equal to or greater than 0.1 MeV, the critical organ is taken to be the whole body and R is set at 0.1 rem/week. If the emissions are alpha particles or beta particles with maximum energy less than 0.1 MeV, the critical organ is considered to be to the skin of the whole body and R is taken to be 0.6 rem/week. Krypton 85 falls in the former group and is considered to deliver its dose to the whole body even though the deposition of energy from externally incident 0.672 MeV beta particles will be deposited within about a 2 mm depth in tissue (11,22) with the average penetration being slightly greater than 0.2 mm (22). The overall effect is to overestimate the dose

[2]The lung is used as the critical organ for radon in equilibrium with its daughters.

actually delivered to the whole body. This will be discussed in detail later. It has been recognized that the MPC values in use are conservative (23,24), and the next issue of the ICRP Recommendations is expected to increase them by a factor of about 5 (24).

^{85}KR AS AN ENVIRONMENTAL CONTAMINANT

HOW AND WHEN ^{85}KR IS RELEASED TO THE ENVIRONMENT

Krypton 85 is produced by nuclear explosions and continuously during reactor operations. It has been concluded (25,26), from considerations of experimentally determined air concentrations versus total weapons yield, that most of the ^{85}Kr in the air, even during a period of active atmospheric weapons testing, is due to reprocessing of reactor fuel. This conclusion is strongly supported by evaluation of the data of Logsdon and Chissler (27) and Kahn *et al.* (28) which shows that about 0.02% of the ^{85}Kr formed in reactor operations from 1959 through 1968 was released to the air at the reactor.[3] The ^{85}Kr produced in reactor fuels is not released, in the absence of cladding failure or "tramp" uranium,[4] until the fuel elements are cut apart in the reprocessing plant and the fuel is dissolved preparatory to chemical separations. Goode (29) reported that 99-99.5% of the release occurs in the dissolution phase with acid treatment of ThO_2-UO_2 fuel in the laboratory. This estimate is confirmed for full scale processing by data of Cochran *et al.* (30) who report that all but about 20-50 curies of the approximately 5,000 curies of ^{85}Kr released per batch processed at the Nuclear Fuel Services plant are released during dissolution. Other radioactive gases released during reprocessing include ^{131}I, ^{129}I, ^{131m}Xe, ^{133}Xe and ^{3}H. Krypton 85 is the only gas, other than ^{3}H, released in sufficient quantity and having a half life long enough to produce significant widespread concentrations in the air. Whipple (50) estimates that ^{85}Kr emission will limit U.S. nuclear power to about 150,000 MW(e).[5]

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[3]Assuming 0.2 kCi of ^{85}Kr produced/MW-year, all beta-gamma activity released to air by PWR/HTGR was ^{85}Kr and 0.001% of total fission gas release from BWR was ^{85}Kr (estimated from isotope composition data in 27,28).

[4]According to Kahn *et al.*(28), the principal sources of fission products in reactor coolant are holes or cracks in cladding or fission of uranium in the coolant that has escaped from failed fuel elements. Tramp uranium and direct diffusion through intact cladding are minor sources.

[5]The release limit by the U.S. power industry was determined to be 10^{10}Ci/year assuming (a) $(MPC)_a$ of 3×10^{-7} μCi/cm^3 reduced by 1/3 for individual variations and 1/10 for summing of dose from several isotopes, (b) half the ^{85}Kr released is from explosion (U.S.), (c) all ^{85}Kr produced is released, and (d) the U.S. uses 0.06 of world capacity (fair share based on population).

Small amounts of ^{85}Kr have been released by venting of cavity gas from certain Plowshare projects such as Gasbuggy and Rulison. Gasbuggy produced an estimated 350 Ci of ^{85}Kr which resulted in a cavity gas concentration of 2.8 $\mu Ci/ft^3$ (31,32). The ^{85}Kr concentration in the cavity decreased exponentially as gas was removed (32-35). The amount of ^{85}Kr released by these tests is minor in comparison to total ^{85}Kr releases, but is of the same magnitude as the amount of ^{85}Kr released at reactor sites through 1968.

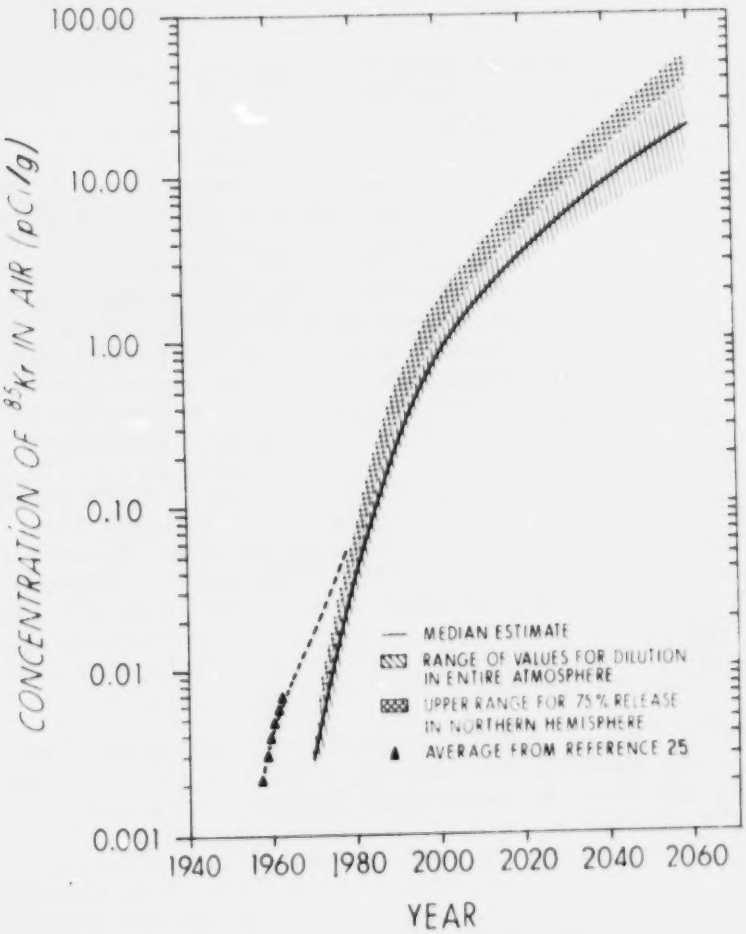

Figure 2. Estimated krypton-85 concentrations in air, 1970-2060.

DISTRIBUTION OF ^{85}KR

WORLDWIDE CONCENTRATION AND DOSE ESTIMATES

Colemen and Liberace (13) estimated future world ^{85}Kr levels, and radiation doses resulting therefrom, based on projected world energy requirements and that part expected to be met with nuclear power. Their estimates, shown in figures 2 and 3, assume that all ^{85}Kr produced is released and that the $(MPC)_a$ actually delivers the permitted dose. Their estimates of air concentrations, published data from a number of laboratories (25,26,36-45), and estimates of the United Kingdom contribution

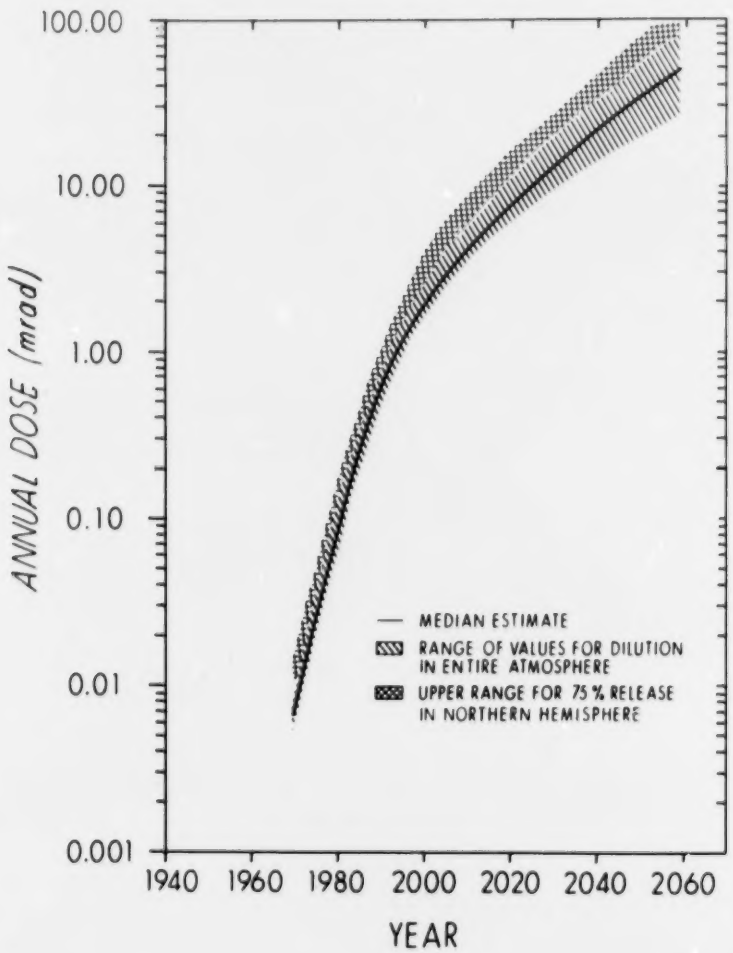

Figure 3. Estimated annual dose from krypton-85, 1970-2060.

(24) were converted to a common unit and are shown in figure 4.[6] Data from Shuping *et al.* (44,45) are shown in figure 5 with the time scale expanded. The ^{85}Kr levels appear to be increasing at a rate near or slightly greater than the predictions. The predictions were not claimed to be accurate in the 1965-1980 period mainly because of uncertainty in the time delays from irradiation to reprocessing.

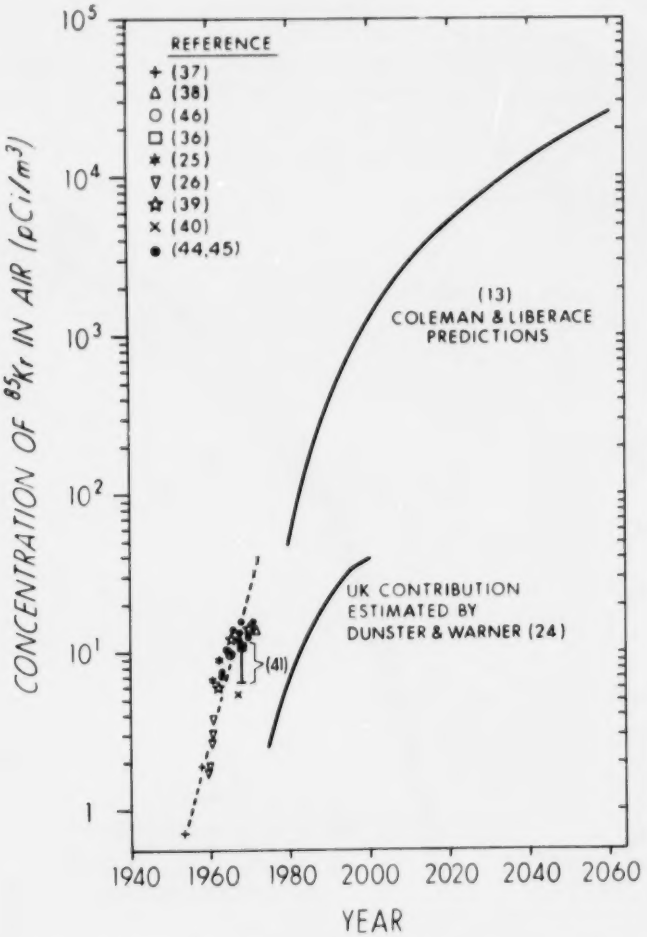

Figure 4. Comparison of estimated ^{85}Kr concentration in air, 1970-2060 with measurements through 1970.

[6] In coverting the U.K. data, the cumulative ^{85}Kr was assumed to be diluted into the total atmosphere of 5.14×10^{21} g at 0.001293 g/cm^3 (3.97×10^{18} m^3) which yields a conversion factor of MCi/3.97 = pCi/m^3.

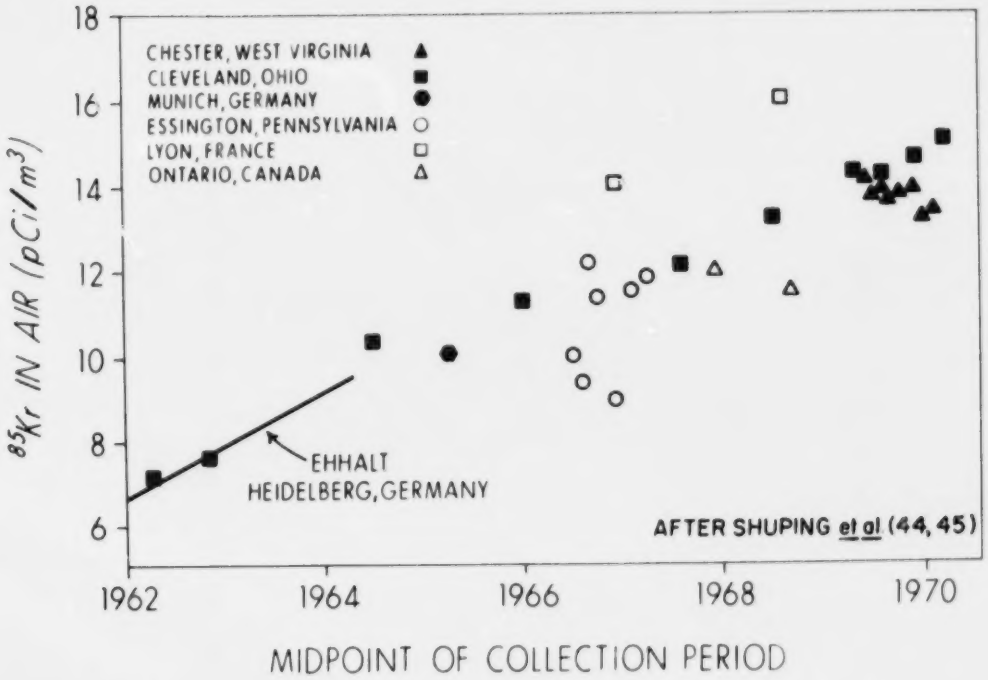

Figure 5. Krypton-85 concentration in air, including data from Ehhalt *et al.* (25).

The data from figure 3 have been replotted in figure 6 for comparison with similar estimates from Dunster and Warner (24), Cowser *et al.* (46,47) and Csongor (38). The U.K. estimates (24) are based on *de novo* dose calculations while the others assume that a dose of 0.5 rem is delivered when the average concentration is 3×10^{-7} $\mu Ci/cm^3$ for one year. The calculations involved are discussed in the section on hazard evaluation.

Krypton 85 in commercial krypton supplies began causing problems as early as 1963 when Ostroski and Jelen (43) reported background problems with krypton-filled ionization chambers. Lasseart and Kellershohn (48) reported similar problems with self-triggering spark chambers in 1965. Dunster and Warner (24) warn of the possibility of personnel hazards associated with handling commercial krypton supplies long before atmospheric concentrations present a significant problem.

For comparison, the cumulative whole-body radiation dose from all nuclear testing conducted through 1962 (end of large scale atmospheric tests) is estimated to be 110 mrem in 30 years (49), which is itself about 1/30 of the dose received from natural sources over the same period (peak dose rates from fallout are greater).

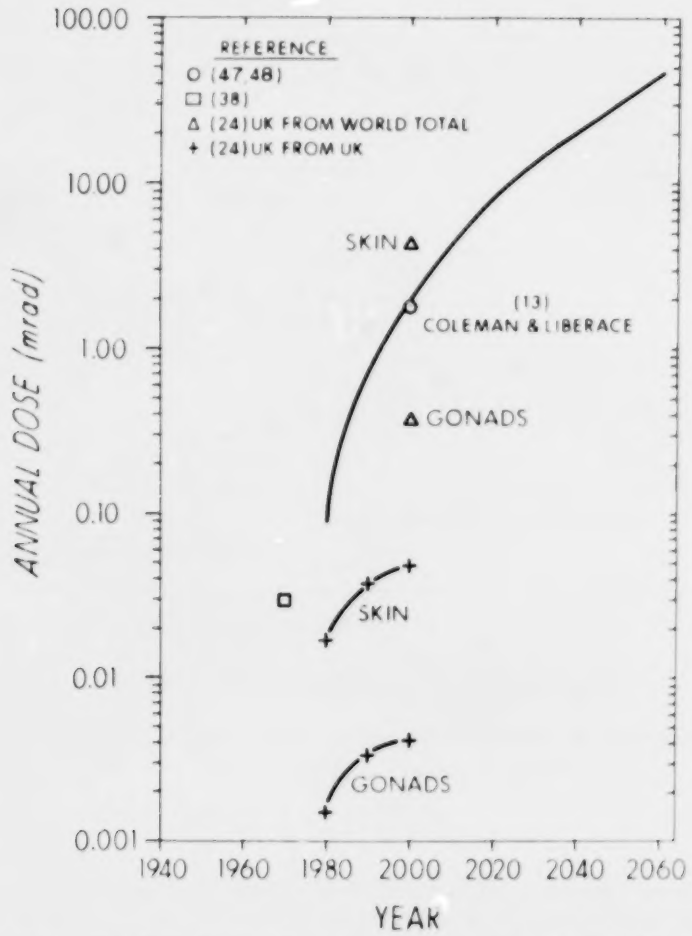

Figure 6. Estimations of annual dose rates from ^{85}Kr.

^{85}KR CONCENTRATIONS AND DOSES NEAR REPROCESSING FACILITIES

Personnel of the Northeastern Radiological Health Laboratory of the Bureau of Radiological Health, PHS, DHEW, have investigated the ^{85}Kr concentrations in the vicinity of the Nuclear Fuel Services reprocessing plant at West Valley, New York, which is the only operating commercial fuel processing facility. These data are reported in references (30) and (51). Concentrations were monitored, as near to the periphery of the NFS property as access roads permitted, during the dissolution of several batches of fuel (about 1 ton/batch). Using data collected in

1968 and early 1969, Shleien (51) reported that the average annual ^{85}Kr concentration at the plant boundary (1.5 km from the stack) would be 2.3 x 10^{-11} $\mu Ci/cm^3$ and that the maximum annual concentration would be 1.3 x 10^{-10} $\mu Ci/cm^3$. These values, derived from plume measurements using wind data and diffusion equations, correspond to annual doses of 0.05 rem and 0.03 rem respectively, if the ICRP values are used to convert the concentrations to doses. In a later report, Cochran *et al.* (30) reported that the ^{85}Kr concentrations in the plume ranged from 1.7 x 10^{-8} to 7.65 x 10^{-7} $\mu Ci/cm^3$ average for a 3-hour dissolution cycle; the peak values during the same time were 1.3 x 10^{-7} to 9.3 x 10^{-6} $\mu Ci/cm^3$. They calculated that the highest annual concentration, 1.7 x 10^{-10} $\mu Ci/cm^3$, would occur at the property line in the north octant. The maximum 24-hour off-property concentration was estimated to be 6.8 x 10^{-7} $\mu Ci/cm^3$. Sax *et al.* (36) reported a concentration of 5.6 x 10^{-10} $\mu Ci/cm^3$ about 5 miles from the plant on one occasion in February 1968.

Dunster and Warner (24) estimate that the dose to individuals in the vicinity of United Kingdom reprocessing facilities in 2000 AD will be about 45 mrad/year to the skin and 0.38 mrad/year to the gonads.

REMOVAL OF ^{85}KR FROM PROCESS STREAMS BEFORE RELEASE TO THE ATMOSPHERE

It seems improbable that overall atmospheric ^{85}Kr concentrations will require corrective or preventive action on purely radiological safety grounds before sometime in the next century (13,24). However, the anticipated growth in size of reprocessing facilities, coupled with increasing cost of enough land to permit MPC to be reached by diffusion before the plume crosses the property line, will probably lead to installation of equipment to remove the ^{85}Kr from the process stream before it leaves the stack. Blomeke and Perona (52) and Perona *et al.*(53) estimate that this point will be reached when more than 5 tons/day of 150-day aged fuel or 0.5 ton/day of 30-day fuel is reprocessed.[7] East Germany requires facilities for storing ^{85}Kr originating in reprocessing plants (54).

Concern with removal of ^{85}Kr dates to the late 1950's (13,55-57). A number of techniques have been investigated including:

1. Adsorption onto activated charcoal at cryogenic temperatures (24,58-61 and many others)
2. Solvent extraction (24,42,55,56,62-64)
3. Condensation in liquid nitrogen followed by fractional distillation (65)
4. Selective permeating through cellulose acetate or silicone rubber membranes (66,67).

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[7]Their analysis included contribution from all noble fission gases, but only ^{85}Kr is important 150 days after cooling.

Three of these techniques have been developed to the operating stage. The solvent extraction method developed at the Oak Ridge Gaseous Diffusion Plant over the past several years (63,64) was recently reported to be commercially available (62). A plant using the LN_2 condensation-redistillation technique is in use at the Idaho Chemical Processing Plant (NRTS) (65). Air and Water News (62) also reports that one commercial reactor supplier is using a charcoal bed removal system.

The most practical disposition for recovered ^{85}Kr appears to be long-term storage in high pressure steel cylinders (24,52,53). Incorporation of ^{85}Kr into glasses, resins, clathrates, molecular sieves, and pressurized steel or glass bulbs in an epoxy matrix have been considered for secondary containment of the ^{85}Kr inside the steel cylinders (68). Serious attention has been given to the possibility of pumping ^{85}Kr into underground storage areas, such as abandoned gas or oil wells or similar formations (52,69,70). This method requires a porous storage formation with an essentially non-porous cap formation that is free of vertical channels. This requirement is too restrictive to permit generalized use at reprocessing plant sites.

One comprehensive proposal, made by Blomeke and Perona (52,53), calls for separating the ^{85}Kr from the process stream, alone or with xenon, by solvent extraction or cryogenic distillation. Processing 2600 tons/year of LWR[a] fuel is estimated to yield 28 cylinders, each containing 50 liters or 10^6 Ci of ^{85}Kr (heat production 5,800 BTU/hour), or 160 cylinders each containing 50 liters of mixed Kr/Xe (180,000 Ci ^{85}Kr, heat production 1000 BTU/hour). The cylinders would be temporarily stored underwater on site and then shipped, in special water-cooled casks, to underground salt mines and stored above the floor in sealed rooms. The storage space requirement for one year's production is one quarter acre and is determined by heat production. The cost of disposal of ^{85}Kr by this technique, including (1) filling, testing, and on-site storage of the cylinders, (2) shipment to a salt mine, and (3) permanent storage in the salt mine, is estimated to be \$190,000-220,000/year for a 2600 ton-year plant. This amounts to 0.0003-0.00035 mills/kW-hr of electricity generated by the reprocessed fuel. This is about 0.001% of the residential electric rate in Rochester, New York in 1971.

Dunster and Warner (24) make basically similar proposals except that they consider solvent extraction or adsorption on activated charcoal to concentrate the ^{85}Kr and evaluated several different types of storage tanks.

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[a]The fuel considered is from a light water reactor (LWR) exposed to 33,000 MWd/ton at 30 MW/ton. An equivalent amount of fuel from a liquid metal fast breeder reactor (mixed core and blanket) with an average exposure of 33,000 MWd/ton at 58 MW/ton will yield about 10% less volume of noble gases.

USES OF KRYPTON 85

MEDICINE AND CLOSELY ALLIED AREAS

Krypton 85 has found important clinical use in the past 15 years. Its physiological characteristics of low blood solubility, high lipid solubility, and rapid diffusion, together with versatility of detection, facilitate differential diagnosis. Specific applications have included:

1. Determination of total body fat (71-75)
2. Circulatory studies
   - a. General (72, 76-86)
   - b. Rate of blood flow
     - (1) Brain
       - (a) Whole and regional (87-116)
       - (b) Partition coefficients (100, 117, 118)
       - (c) Detection of lesions (87, 102, 119)
     - (2) Heart
       - (a) Output (120-125)
       - (b) Myocardial flow (126-132)
     - (3) Lungs and perfusion (133-140)
     - (4) Kidneys (108, 141-146)
     - (5) Skin (147-149)
     - (6) Gastric mucosa (150-152)
     - (7) Intestines (153-155)
     - (8) Liver (156,157)
     - (9) Eyes (158, 159)
     - (10) Muscle (106, 160)
     - (11) Testis (161)
     - (12) Tumors (162)
     - (13) Fresh grafts (163)
   - c. Circulatory shunts
     - (1) Left-to-right, including atrial septal defects, ventricle septal defects, and patent ductus (164-173)
     - (2) Right to left (pulmonary) (168, 174-179)
     - (3) Hepatic-pulmonary (177)
     - (4) Hepatic to vena cava (180)
     - (5) A-V aneurisms in brain (117)
3. Lung function studies - emphysema, cysts, cancer etc. (133-140, 181-183)
4. Structure of teeth (crystalline) (184)
5. Determination of surface area of elastin (185)

The quantities of ^{85}Kr used in these studies have usually been in the μCi-mCi range.

NON-MEDICAL USES OF ^{85}KR

The non-medical uses of ^{85}Kr can be divided into two areas: (1) those that use ^{85}Kr as the gas and (2) those that incorporate it into solids prior to use. Examples of the first category are:

1. Replacement of radium bromide as an ionization source in cold cathode gas discharge tubes (186)
2. Location of carbon monoxide leak into aircraft cabins in flight (187)
3. Measurement of stream aeration (188)
4. Determination of surface area of atmospheric particulates (189)
5. Measurement of gas flow in piping systems (190)
6. Tests of gaseous diffusion theory in solids (68, 69, 191)
7. Measurement of stack gas dispersion and diffusion (31, 192-197)
8. Study of interhemispheric atmospheric mixing (39)
9. Study of other atmospheric gases (40, 41)

Krypton 85 has been incorporated into or onto solids using techniques recently reviewed by Balek (5) and by Eddy (198). Methods include: (1) fission recoil, (2) bombardment of surfaces with high-energy ^{85}Kr ions, (3) diffusion into crystal lattices at high temperatures and pressures (successful with over 150 materials), (4) crystallization of solids from melt or by sublimation in an ^{85}Kr atmosphere and (5) by adsorption onto outgassed surfaces. All but the last method yield more or less stable products which are called kryptonates if surface labeled, as by the first three methods, or clathrates if the ^{85}Kr is incorporated throughout the material as by the fourth method.

The distribution of ^{85}Kr can be determined by autoradiography, or its electronic equivalent, and used to study structural features of solid materials including surface phenomena, lattice structure, or channeling (5, 199-201). Cracks and imperfections in machinery components, such as turbine blades, can be detetected after either kryptonation or adsorptive labeling. Autoradiography is the only technique sensitive enough to use with the kryptonated materials while either autoradiography or electronic imaging works well with the adsorptive technique.

Another potentially extensive use of kryptonates and krypton clathrates lies in the field of chemistry (4, 5, 202). Krypton 85 can be incorporated into one of the reactants, catalysts, or incidental materials, and the release of the gas from the solid used to detect the beginning of a reaction or to measure its rate. Reaction end points, such as in titrations, can be objectively determined by including in the solution a kryptonated solid that will not react until an excess of titrant is present.

The amount of ^{85}Kr involved in non-medical uses is rarely mentioned; however, some of the counting data presented and description of the procedures suggest that some procedures may use curie quantities.

RADIATION HAZARDS ASSOCIATED WITH ^{85}Kr

SKIN DOSE

The dose to the skin from a cloud of ^{85}Kr is the sum of doses from beta particles, gamma rays and Bremsstrahlung from the surrounding atmosphere and from the ^{85}Kr that has been absorbed into the body. In the circumstances postulated in deriving the $(MPC)_a$, only the contribution from outside the body is significant; the internal contribution will be 1-2 orders of magnitude less than the external gamma/Bremsstrahlung component and 4-5 orders of magnitude less than the dose delivered to the skin by external beta radiation.

The surface beta dose in an infinite cloud of a beta-emitting gas such as ^{85}Kr is given by:

$$D = 1.07 \times 10^{-6} C_a \bar{E} K \text{ rad/hour} \quad (24)$$

where: C_a = pCi ^{85}Kr/gram of air
$\bar{E}$ = average beta energy in MeV = 0.249 MeV[9] (11)
K = ratio of stopping power in tissue to stopping power in air = 1.15 (24)

This reduces to:

$$D = 3.064 \times 10^{-7} C_a \text{ rad/hour} = 2.68 \times 10^{-3} C_a \text{ rad/year}$$

or

$$D = 2.07 C_a' \text{ rad/year where } C_a' = \text{pCi } ^{85}Kr/cm^3 \text{ air}^{10}$$

This equation represents 50% of the point dose at the center of an infinite cloud of ^{85}Kr or the dose at the surface of an infinite slab of ^{85}Kr, multiplied in either case by the stopping power ratio.

The expression for the dose to the surface of the body from gamma radiation and Bremsstrahlung was derived by N. Adams in Appendix 1 to reference (24). This dose is given by the expression:

$$D = 2.42 \times 10^{-9} C_a \text{ rad/hour} = 2.12 \times 10^{-5} C_a \text{ rad/year}$$

or

$$D = 1.64 \times 10^{-2} C_a' \text{ rad/year}$$

where C_a = pCi ^{85}Kr/gram of air and C_a' = pCi $^{85}Kr/cm^3$ of air as above.

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[9] Reference (24) uses 0.234 MeV. Results differ accordingly.
[10] Using 1 cm^3 air = 0.001293 gram air.

DOSE IN THE BODY

DOSE FROM ^{85}KR OUTSIDE THE BODY

The average dose to the total body from gamma radiation and Bremsstrahlung from ^{85}Kr outside the body (infinite cloud) is given by (24-Appendix 1):

$$D = 1.97 \times 10^{-9} C_a \text{ rad/hour} = 1.73 \times 10^{-5} C_a \text{ rad/year}$$

or

$$D = 1.38 \times 10^{-2} C_a' \text{ rad/year}$$

DOSE FROM ^{85}KR CONTAINED IN THE BODY

To calculate the dose of ^{85}Kr in the body that results from a given concentration of the isotope in the surrounding air, the fraction of the air concentration that will ultimately be found in the body, or a specific part thereof, must be known. This faction is the partition coefficient which is usually designated as λ.

The internal behavior of ^{85}Kr is discussed in detail in Appendix A, including gas solubility, partition coefficients, and kinetic parameters. The partition coefficient for the body or a particular tissue is largely dependent on its fat content and for ^{85}Kr is closely approximated by:

$$\lambda \text{ tissue:air} = 0.06 \frac{(9V_f + V_r)}{V_t} = (0.48V_f/V_t) + .06$$

where: V_t = total tissue volume, $V_t = V_f + V_r$
V_f = volume of fat in tissue
V_r = volume of rest of tissue

Thus the partition coefficients from body tissue to air ranges from 0.06 to 0.54. The average partition coefficient for standard man is 0.163

To calculate the tissue concentration of ^{85}Kr at equilibrium, the air concentration is multiplied by the partition coefficient. If tissue density is unity, the result is in pCi/gram.

The dose which results from the gamma and Bremsstrahlung component of ^{85}Kr in the body is given by (24, Appendix 1):

$$D = 1.43 \times 10^{-9} C_t \text{rad/hour} = 1.25 \times 10^{-5} C_t \text{rad/year}$$

where C_t = pCi ^{85}Kr/gram of tissue ($C_t = C_a' \times \lambda$)

The beta component of internal dose is given by:

D = C_tpCi/g x 2.22 disintegrations/pCi-min x 0.9959 beta/disintegration x 0.249 MeV/beta x 1.6021×10^{-6} ergs/MeV x rad-g/100 ergs x 60 min/hour
= 5.29×10^{-7} $C_t$rad/hour = 4.62×10^{-3} $C_t$rad/year

If λ air:tissue = 0.06; D = 2.77×10^{-4} C_a' rad/year
λ air:tissue = 0.163; D = 7.58×10^{-4} C_a' rad/year
λ air:tissue = 0.54; D = 2.50×10^{-3} C_a' rad/year

where C_a' = pCi $^{85}Kr/cm^3$ in the surrounding air.

DOSES TO SKIN, WHOLE BODY AND MALE GONADS AT UNRESTRICTED $(MPC)_a$

An individual continuously submerged in an infinite cloud of ^{85}Kr at a concentration of 3×10^{-7} $\mu Ci/cm^3$ (C_a' = 0.3 pCi/cm^3, with an air density of 0.001293 g/cm^3 C_a = 232.56 pCi/g air) receives the following annual doses.

SKIN (SURFACE)

External beta dose = 2.073 C_a' rad/year = 0.623 rad
Gamma and Bremsstrahlung dose = 1.64×10^{-2} C_a' rad/year = 0.00493 rad
Total = 0.628 rad

GONADS (MALE)

Gamma and Bremsstrahlung dose at surface =
1.64×10^{-2} C_a' rad/year = 4.93×10^{-3} rad

Gamma and Bremsstrahlung dose from ^{85}Kr inside the body (λ = .06) =
1.25×10^{-5} C_t rad/year = 7.5×10^{-7} C_a' rad/year = 2.25×10^{-7} rad

Internal beta dose (λ = .06) =
2.77×10^{-4} C_a' rad/year = 8.33×10^{-5} rad
Total 5.01×10^{-3} rad

WHOLE BODY (λ = 0.163)

Gamma and Bremsstrahlung dose from ^{85}Kr outside the body =

= 1.38×10^{-2} C_a' rad/year = 4.02×10^{-3} rad

Gamma and Bremsstrahlung dose from ^{85}Kr inside the body =

$1.25 \times 10^{-5}\ C_t$ rad/year =

$2.04 \times 10^{-6}\ C_a'$ rad/year = 6.12×10^{-7} rad

Internal beta dose =

$4.62 \times 10^{-3}\ C_t$ rad/year =

$7.58 \times 10^{-4}\ C_a'$ rad/year = 2.27×10^{-4} rad

Total 4.25×10^{-3} rad

OTHER DOSE CALCULATIONS

Depth-dose calculations presented by Hendrickson (203, 204) illustrate the discrepancies between the surface dose and the actual dose in several critical tissues (ignoring contributions from ^{85}Kr in the body). The results of these calculations, which include contributions from beta, gamma, and Bremsstrahlung, for an ^{85}Kr concentration of 3×10^{-7} $\mu Ci/cm^3$ are given in table 1.

TABLE 1.[a] ANNUAL DOSE FROM IMMERSION IN AIR WITH A CONCENTRATION OF 3×10^{-7} $\mu Ci(^{85}Kr)/Cm^3$

Tissue	Tissue Depth (mm)	β Radiation (rem/yr)	x and γ Radiation (rem/yr)
Whole Body	50	Nil	0.007
Gonads	10	Nil	0.007
Gonads	2	4×10^{-7}	0.007
Surface of Skin (or clothing)	0.0	0.5	0.007
Skin (shallowest layer of live skin)	0.07	0.3	0.007
Lens of Eye	2	4×10^{-7}	0.007
Lung	(Internal Surface of Lung)	0.005[b]	0.007

[a]From Hendrickson (203).
[b]Internal exposure to surface lung tissue from ^{85}Kr in the lung.

Detailed calculations of doses resulting from immersion in infinite clouds of the various reactor-produced noble gases, including ^{85}Kr, using the MIRD[11] methodology are nearing completion and will be published in the near future (205).

Relationships for calculating doses to various organs resulting from inhalation or injection of ^{85}Kr during the medical diagnostic procedures previously mentioned have been derived by Lassen (206) who estimates that the following doses would result from breathing ^{85}Kr at a concentration of 1 $\mu Ci/cm^3$ for 1 minute:

Tracheal mucosa	- 71.9 mrad
Lungs	- 27.3 mrad
Adipose tissue	- 4.2 mrad
Other (incl. gonads)	0.5 mrad

These values would change relative to each other as exposure time increases. The original paper should be consulted for details.

COMPARISON OF DOSES DELIVERED AT $(MPC)_a$ WITH PERMITTED DOSES

ICRP 9 (207) summarizes dose limits for occupational exposure and exposure to members of the public as shown in table 2. An additional limit is the recommended whole population genetic limit of 5 rems/30 years (0.167 rem/year).

TABLE 2[a]. SUMMARY OF DOSE LIMITS FOR INDIVIDUALS

Organ or Tissue	Maximum Permissible Doses for Adults Exposed in the Course of Their Work (rems in a year)	Dose Limits for Members of the Public (rems in a year)
Gonads, red bone marrow, whole body	5	0.5
Skin, bone, thyroid	30	3
Hands and forearms, feet and ankles	75	7.5
Other single organs	15	1.5

[a]From ICRP-9 (208).

[11]Medical Internal Radiation Dose Committee of the Society of Nuclear Medicine.

Figure 7 shows the fractions of applicable limits delivered by ^{85}Kr at a concentration of 3×10^{-7} $\mu Ci/cm^3$, to the skin, gonads, and whole body. The full bar in each case represents the fraction given in an infinite cloud with exposure time long with respect to body saturation time. The shaded area of the bar represents the fraction delivered without the external gamma/Bremsstrahlung component. The numbers in parentheses are the factor by which the $(MPC)_a$ is conservative for each case. It appears that the $(MPC)_a$ is conservative by a factor of at least 4.8 and, when shielding effects of clothing and buildings are considered, probably by a much larger factor. Modifying factors of particular importance in the occupational situation are heavy clothing, such as lab coats, and the absence of an infinite cloud with respect to gamma radiation. For example, if 50 mg/cm^2 of clothing is worn reasonably close to the body, the skin

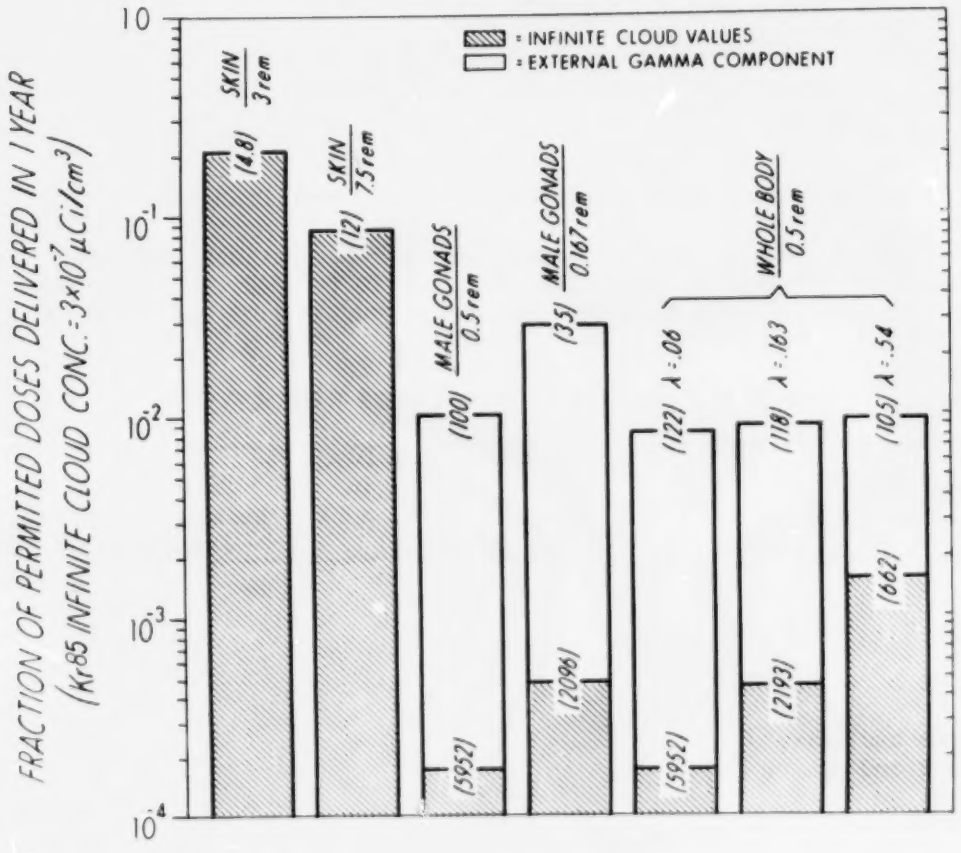

Figure 7. Comparison of estimations of annual dose rates from ^{85}Kr.

beta dose would be 18% of the unclothed dose so the total skin dose/year at 3×10^{-7} $\mu Ci/cm^3$ would be 0.112 rad beta + 0.005 rad gamma or 0.117 rad/year.[12] This is conservative by a factor of 25.6. The controlling dose would then be the extremity dose which is conservative by a factor of 7.5/0.628 = 12.

An increase in the unrestricted $(MPC)_a$ by a factor of 5 and the occupational $(MPC)_a$ by a factor of 10-12 would appear justifiable if the only consideration is meeting existing dose limits, but may not be desirable when other things are considered.

UNEXPLAINED PHENOMENA INVOLVING "INERT" GASES

A number of inert gas phenomena, reported in the literature, have not been satisfactorily explained. These have all occurred at pressures many times greater than those considered in radiation protection, but may be applicable to the physiological behavior of ^{85}Kr and must be considered. The relative mangitude of effects found with members of the helium series usually have been in order of polarizability or oil solubility which is Xe > Kr > A > Ne > He.

The growth of *Neurospora crassa* was inhibited by 50% with 0.8 atm. of Xe, 1.6 atm. of Kr, 3.8 atm. of A, 35 atm. of Ne and 300 atm. of He (208); the inhibition of growth at 650 mm pressure was found to be proportional to the square root of the atomic weight of the gas.

High pressures of xenon (225 psi) led to cessation of motion, a decrease in contractile vacuole activity, increase in cell volume and surface area and cytoloysis in paramecia (209). Krypton produced a decrease in contractile vacuole activity at 915 psi.

Reversible inhibition of Na^+ transport across frog skin was found at 200 psi xenon or 950-1000 psi krypton (210). Similar pressures caused reversible blockage of frog sciatic nerve transmission. In similar frog nerve-muscle preparations, 100 psi xenon caused a decrease in muscle contraction and nerve conduction and 200 psi krypton decreased muscle contraction (211).

Radioprotection by inert gases is reported for animals (212) and bean sprouts (8) although krypton had no protective effect on mice at 2 atmospheres pressure (213). Markoe *et al.* (214) reported potentiation of killing of HeLa cells by x rays which was proportional to the partial presure of Xe or Kr present at irradiation.

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[12]Since the limits for members of the public are each 1/10 of the corresponding occupational limit, the degree of conservativeness calculated for one applies to the other.

Possibly the most interesting mystery is the demonstrated anesthetic action of xenon (6) in humans. Surgical procedures (orchidectomy and Fallopian tube ligation) have been performed using an 80% Xe:20% O_2 mixture (1 atm. total pressure) for anesthesia. Higher partial pressures of Xe are required to anesthetise animals. The narcotic potency of the helium series seems to follow the order previously stated (215) although narcotic effects of krypton are equivocal at atmospheric pressures (6).

Another observation that has not been accounted for is the uptake of 1 1/2 - 2 times as much inert gas by the adrenal as by any other tissues (8).

The foregoing phenomena are not understood at this time although they have been widely debated (3, 7, 8). The most popular postulates, particularily for anesthesia, are membrane effects involving lipid solubility or stabilization of the formation of hydrate microcrystals in the nervous system to block electrical conduction.

SAMPLING AND ANALYSIS OF ^{85}KR

DETECTION AND COUNTING

GENERAL

Krypton 85 has been successfully detected and counted by virtually every conventional beta or gamma detection technique, and some unconventional ones, when sampling methods and counting techniques have been properly matched with the amount and concentration of isotope to be analyzed and its physical form or configuration.

DETECTORS USED

GM Counters: GM counting has been used extensively in the medical studies cited. End window GM detectors were used to count the beta radiation emitted from the surface of organs, thin-windowed sample containers, or flowing gas mixtures. Internal GM counters have been used to analyze environmental samples (43).

The detection efficiency of end-window GM tubes is determined by many factors and may approach 50% with good geometry and thin windows. Martin (192) showed that commercial GM tubes with 30 mg/cm^2 walls will detect 5 x 10^{-7} μCi/cm^3 of ^{85}Kr in an infinite cloud geometry. Ludwick *et al.* (194), using a very sensitive tube with a 3.5 mg/cm^2 window 50 mm in diameter, were able to detect the passage of a cloud with a concentration of 10^{-7} μCi/cm^3. GM tubes coupled to ordinary survey meters have been successfully used to assay the concentration of ^{85}Kr in

pressurized steel cylinders (216). Most recently, Smith *et al.* (217) report calibration of several types of GM detectors to ^{85}Kr in a known fraction of an infinite cloud. Their results are shown in table 3.

GM tubes are commonly used as stack monitors by placing them in a tank large enough to provide an appreciable fraction of an infinite cloud (for beta) and drawing part of the stack flow through the tank (30, 217).

Internal Ionization Counters and Proportional Counters: The standard technique of counting radioactive gases in internal proportional counters or ionization chambers has been used by several authors to analyze ^{85}Kr in environmental samples (25, 31, 217-219) and is the method used in standardization. Gas flow proportional counting was used in conjunction with gas-phase chromatography by Dupuis *et al.* (220). Calibration data reported by Smith *et al.* (217) for various sized ionization chambers is shown in table 4. These same chambers were equipped with needle valves and flowmeters and used to obtain and count integrated atmospheric samples near the Nuclear Fuel Services plant by Cochran *et al.* (30). Cold trapping of xenon and radon was required.

Ionization chambers may be used in the flow-through mode as well as a static mode. This method was used in characterizing the ^{85}Kr concentration used in calibrating GM tubes to infinite cloud geometry (217).

Scintillation Counters: Gamma Scintillation - Next to GM counting, gamma scintillation counting using NaI(Tl) crystals is used most to assay ^{85}Kr. The 0.514 MeV gamma photon emitted in about 0.43% of the ^{85}Kr disintegrations penetrates tissue or sample containers easily and can be detected with the NaI(Tl) crystal. This has been the technique used in most of the papers reporting whole-body or whole-organ saturation or desaturation data and in laboratory analyses if the activity was high enough. The technique avoids the sample self absorption problems encountered with GM counting and the preparation problems involved with most of the following techniques. However, the low detection efficiency of the NaI combined with the low gamma emission rate of the ^{85}Kr results in very low efficiency.

Beta Scintillation - Both liquid and solid beta scintillation techniques have been used with ^{85}Kr. Liquid scintillation has become an increasingly popular method of counting ^{85}Kr and takes advantage of the high solubility of Kr in toluene based scintillation cocktails (44, 45, 221, 222). Very low specific activity samples may require concentration by cryogenic techniques before counting. According to Shuping *et al.* (44, 45), the lowest concentration that can be analyzed without preconcentration is about 3 pCi/ml. Counting efficiency is 92-100% and 0.014 pCi of ^{85}Kr can be counted with good precision. The limiting problem with air mixtures is the poor solubility of the air in the scintillator. The undissolved air forms a pocket and part of the Kr goes out of solution into the air.

TABLE 3. MINIMUM DETECTABLE ^{85}KR CONCENTRATIONS FOR CALIBRATED EXTERNAL β DETECTORS [after Smith *et al.* (217)]

Count Time Background/Sample Detector Type (Model)	Laboratory MDC (pCi/cm^3)		Field MDC (pCi/cm^3)	
	Long Count[a] 0.5/4 hr	Short Count[b] 10/10 min	Long Count[c] 0.5/4 hr	Short Count[d] 10/10 min
2 window pancake GM (Eon 8008H)	.007	.012	.020	.012
1 window pancake GM (Amperex 18546)	.011	.024	.042	.027
(Eon 8001T)	.014	.025	.040	.025
Cylindrical Probe (LND 719)	.011	.024	.043	.024
(Eon 5108E)	.013	.024	.046	.028
β Scintillator (Pilot B)	.016	.029	.045	.029

Notes on counting conditions:

(1) All values assume the MDC = $2s/C_{85}$ where s = the standard deviation of the measurement (CPM) and C_{85} = the calibration factor for the detector (CPM/(pCi/cm^3)).

(2) Total instrument errors are assumed to be negligible in each case.

(3) The magnitude of the relative background variations ($2s_{vb}/B$, where B = background CPM and s_{vb} = standard deviation due to background level fluctuations) assumptions for the four counting interval combinations used are denoted by the following:

Superscript	Relative background variations ($2s_{vb}$)
a	4.8%
b	9.8%
c	27.4%
d	11.8%

(4) 0.3 pCi/cm^3 $(MPC)_a$ for individuals in the general population, 10CFR20 (19).

TABLE 4. MINIMUM DETECTABLE CONCENTRATIONS FOR ^{85}KR IN IONIZATION CHAMBERS[a,b]

Chamber volume (liters)	MDC ^{85}Kr Chamber Unshielded (pCi/cm^3)	MDC ^{85}Kr Chamber/2" Pb Shields (pCi/cm^3)
0.5	1.3×10^{-1}	1.5×10^{-1}
1.0	1.9×10^{-1}	1.5×10^{-1}
2.8	3.9×10^{-2}	3.1×10^{-2}
4.3	3.9×10^{-2}	2.3×10^{-2}

[a]Cary-Tolbert design (Applied Physics Corp.).
[b]After Smith *et al.* (217).

An ingenious counting technique, reported by Sax *et al.* (223) for environmental samples, employs cryogenic preconcentration by 5 Å molecular sieves at liquid nitrogen temperatures and counting tubes full of plastic scintillator shavings. The shavings are outgassed under vacuum and the concentrated sample is drawn into the tube by vacuum. The tubes are reusable after outgassing.

Smith *et al.* (217) also tested Pilot B scintillator for uses similar to those of GM tubes and found that it was less sensitive than the GM detectors (see table 3).

A pressurized scintillation chamber is described by Voice (224).

Semi-conductor Detectors: Semiconductor detectors of both the lithium-drifted pi-n (108) and the silicon p-n (225) types have been used in inert catheters for *in vivo* monitoring of ^{85}Kr concentrations in the heart and lungs.

Integrating Dosimeters: Thermoluminescent dosimeters and film badges have both been used for monitoring ^{85}Kr (226). The major drawback with both is difficulty in characterizing the exposure as to radiation type and energy to properly assess dose.

CALIBRATION AND STANDARDIZATION

Krypton 85 gamma or beta calibration standards are available from the National Bureau of Standards or other suppliers. Secondary ^{85}Kr standards can be prepared by comparison. Gamma calibration has been done with ^{85}Sr sources, an adequate procedure when only the 0.514 MeV photopeak is used. The Bremsstrahlung spectrum from ^{85}Kr overwhelms the Compton shelf at lower energies and precludes using ^{85}Sr without a lower discriminator. Also ^{198}Au has been used as a counting standard (227) but is not recommended. GM tubes were calibrated to measure ^{85}Kr beta radiation by immersion in a ^{204}Tl solution (228). Data comparing the response to ^{85}Kr in air and calibration factors are given.

SAMPLING AND SAMPLE PREPARATION

The gaseous nature and relative chemical inertness of ^{85}Kr preclude using the normal concentration techniques of drawing air through a filter or an activated charcoal cartridge at ambient temperature. With sufficient specific activity, an appropriate sample container can be filled and counted by any of the mentioned techniques, or a flow-through sampling and counting technique can be used with ionization or proportional counters. For samples of very low specific activity such as those collected in the environment at large, or samples containing other radioactive inert gases, concentration and/or separation by cryogenic techniques are the usual procedures. Molecular sieves (223, 229), activated charcoal (227, 229, 230), silica gel, vermiculite and alumina (229) have been successfully used as collectors with liquid nitrogen cooling. Copper wool was used successfully in a LN_2 cooled cold trap to collect Xe (231) and was suggested for collection of Kr. Gas chromatography is becoming popular for analysis of ^{85}Kr in the presence of other noble fission gases (220, 232-234). Direct condensation in LN_2 or LOX, followed by redistillation has been used successfully (26, 64).

In theory, any of the separation techniques discussed under removing ^{85}Kr from the air can be used. The ones not mentioned in this section have practical drawbacks such as bulk or less than 100% recovery. All of the adsorbing media have a finite capacity. The quantity of media used and the operating temperature must be matched to the flow rate and the sample size required, and the system must be tested in its designed service before field use is attempted.

SUMMARY

Krypton 85 is an environmental contaminant for which progress in development of monitoring and control methodology appears to have outstripped knowledge and understanding of its biologic effects. Monitoring methods presently available, while subject to improvement, are adequate for routine use and several reasonable techniques for removing ^{85}Kr from effluent gas streams have been demonstrated on at least pilot plant scale. On the other hand, the value judgments regarding the necessity for and stringency of control of release have been and are being based on calculations and extrapolations rather than on experimental data obtained with living systems. Data on the effects of both acute and chronic exposure of several animal species to ^{85}Kr are needed to confirm these extrapolations.

Until the behavior and effects of radioactive noble gases in living systems are better understood, the basis for release regulations will continue to be founded entirely on radiation dose calculations. These calculations should be subjected to the scrutiny of *in vivo* experimentation. In the interim, control of the major source of ^{85}Kr release, fuel reprocessing plants, to the lowest practical emission level is desirable. It may be appropriate, especially if the $(MPC)_a$ for unrestricted areas is relaxed, to require these plants and any other major source that may develop to control their release to levels much lower than necessary to reach the $(MPC)_a$ at plant boundaries.

Relaxation of the occupational $(MPC)_a$, albeit justifiable by calculations, is subject to the same reservations. It appears more desirable to grant exceptions or modifications in individual cases for variations in exposure conditions, such as wearing of heavy clothing or absence of an infinite cloud, than to change the standard at this time.

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APPENDIX A

ABSORPTION OF KR INTO THE BODY

1. *SOLUBILITY OF ^{85}KR IN VARIOUS MATERIALS: PARTITION COEFFICIENTS*

(General references - 1,2)

Gas solubility data in the literature are frequently expressed in terms of either the Bunsen solubility coefficient (α) or the Ostwald solubility coefficient (L). In component units:

$$\alpha = \frac{Vo(a)}{V} \frac{Po}{P}$$

where: Vo(a) = volume of gas absorbed
V = volume of absorbing fluid
Po = 760 mm Hg
P = partial pressure of the gas being absorbed in mm Hg

and $L = \alpha\, T/To$

where: To = 273° K
T = 273° K + temperature at which gas is absorbed in °C

The Ostwald coefficient, L, is also commonly called the partition coefficient and is sometimes designated as λ. As commonly used, a partition coefficient will be written as λ medium 1:medium 2 (example λ blood:air) and is the ratio of the volume concentration in medium 1 to the volume concentration in medium 2 at equilibrium. If λ blood:air = 0.06, and the air concentration is 1 μCi/cm^3, the blood concentration will be 1 μCi/cm^3 x 0.06 = 0.06 μCi/cm^3.

The solubility of Kr has been determined *in vitro*, usually with degassed solvents, for a number of solvents and solutions. These data are summarized in Table A-1.

2. *IN VIVO PARTITION COEFFICIENTS*

The tissue:air partition coefficient (λ) for an organ or system is given by:

where: λ tissue:air = Σ (λ_i:tissue:blood x f_i) λ blood:air
λ_i = tissue:blood partition coefficient for component i
f_i = the fraction of the organ or system made up of component i.

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TABLE A-1. SOLUBILITY COEFFICIENTS FOR ^{85}KR IN VARIOUS SOLVENTS[a]

Solvent	Temp(°C)	Bunsen Coefficient	(ref)	Ostwald Coefficient	
Olive oil	45	0.3844	(13)	0.4477	(13)
	37	0.4031	(13)	0.4581	(13)
		0.43	(35)		
		0.44	(36)		
	30	0.4225	(13)	0.4688	(13)
	25	0.4376	(13)	0.4746	(13)
	22	0.44	(35)		
	45	0.0441	(14)		
Water	45	0.0441	(14)		
	37	0.0481	(14)	0.0499	(38)
		0.045	(37,35)		
	30	0.0539	(14)		
	25	0.0581	(14)		
	22	0.059	(35)		
Saline soln.	45	0.0411	(14)		
	37	0.0444	(14)		
	30	0.0499	(14)		
	25	0.0542	(14)		
β-albumin	37	0.0195	(12)		
	30	0.0412	(12)		
	25	0.0624	(12)		
Hemoglobin soln. (15.4%)	37	0.0247	(12)		
Blood	37	0.0455	(12)	0.0517	(12)
				0.06	(39)
Muscle homogenate	37	0.0439	(12)		
	30	0.0501	(12)		
	25	0.0549	(12)		
Brain homogenate	37	0.0454	(12)		
	30	0.0517	(12)		
	25	0.0572	(12)		

Solvent	Temp(°C)	Bunsen Coefficient	(ref)	Ostwald Coefficient	
Human fat	45	0.3878	(13)	0.4516	(13)
		0.3875	(13)	0.4513	(13)
	37	0.4071	(13)	0.4626	(13)
		0.4062	(13)	0.4617	(13)
		0.420	(36)	0.425	(41)
		0.414	(36)		
	30	0.4258	(13)	0.4725	(13)
		0.4247	(13)	0.4713	(13)
	25	0.4412	(13)	0.4816	(13)
		0.4404	(13)	0.4807	(13)
Dog fat	45	0.3853	(13)	0.4426	(13)
	37	0.4031	(13)	0.4581	(13)
	30	0.4225	(13)	0.4721	(13)
	25	0.4364	(13)	0.4764	(13)
Rat fat	45	0.3847	(13)	0.4481	(13)
	37	0.4037	(13)	0.4588	(13)
	30	0.4219	(13)	0.4755	(13)
	25	0.4363	(13)	0.4762	(13)
Toluene base liquid scint. cocktail	-15	0.9	(40)		

[a] Addtional ^{85}Kr solubility data in a wide variety of chemicals may be found in references (2, 42-46).

If the tissue components, their relative fractions, and their Ostwald coefficients are known, the tissue:air coefficient can be calculated. In practice this is rarely the case and the coefficient is either determined for the tissue by a dual isotope method such as that of Glass and Harper (3), or, in many cases, estimated to be the same as that for blood. A partial listing of the experimentally determined tissue:air or tissue: blood partition coefficients for ^{85}Kr follows:

Tissue	Part	Tissue:Blood	Tissue:Air	Reference
Brain	Cortex	0.92 (Hc=50)		4,5
	White	1.26 (Hc=50)		4,5
Kidney	Cortex	1.0		6
		0.96		7
	Medulla	1.0		6
Fat		9		6
			0.54	8
		5		5
Liver		1.06		9
		1.04		5
Eye	Retina	1.0		5
Skeletal muscle		1.0		5
Testis		0.85		5,10
Blood			0.05-0.06	Various

Because the coefficients are so similar, Lassen (8), one of the pioneers in this work, normally uses a tissue:air coefficient of 0.06 for all tissues except fat for which he uses 0.54 (9 x 0.06). The only significant variable, for most purposes, is the fat content of the tissue or body being studied. Lassen (8) uses a whole-body tissue:air coefficient of 0.11 for lean people, 0.20 for normal weight people, and 0.30 for obese people. The calculated coefficient for standard man is 0.163.

Whole-body partition coefficients and kinetic parameters for female Rochester Wistar rats, weighing about 250 grams, were experimentally determined by saturating the animals with ^{85}Kr in a closed exposure system for 12 1/2 or 33 1/2 hours, then whole-body counting in a 3- x 5-inch NaI well crystal until the count reached background (about 12 hours). The average seven determinations on four animals was 0.0921 (S. D. = 0.0188). The coefficient for one animal that died during the 12 1/2 hour run, determined by counting component pieces, was 0.0958. The value predicted for rats of 320 grams, using the tissue composition data of Caster, *et al.*(11), and the solubility data of Yeh and Peterson (12, 13, 14) was 0.076. Using Lassen's coefficients (8) and the Caster fat fraction of 0.0708 of the body weight, a value of 0.094 would be predicted.

3. KINETICS OF ^{85}KR ABSORPTION AND DESORPTION IN THE BODY

The rate of absorption of ^{85}Kr into the tissues of the body during exposure and desorption after exposure or injection of the isotope is a complex function of the tissue:blood partition coefficients and, particularly, the blood circulation in the tissue in question. The curves obtained will have as many exponential components as there are differently perfused elements in the tissue being studied. Kety (1) has discussed the kinetics involved in detail. For a single tissue, with the concentration in alveolar air considered constant and blood-tissue diffusion time considered negligible (actually less than 1 sec), the following equations apply:

$$\text{Saturation:} \quad C_i = \lambda_i C_a' (1 - e^{-kt})$$

$$\text{Desaturation:} \quad C_i = C_{i(o)} e^{-kt}$$

where: C_i = concentration in tissue i at time t, $\mu Ci/cm^3$
λ_i = tissue:air partition coefficient
C_a' = concentration in air, $\mu Ci/cm^3$
$k = F_i/V_i$ where F_i = blood flow rate in tissue i, and V_i = volume of tissue i

The quantity of gas in the tissue $(Q_i) = V_i C_i$ or $Q_i = V_i \lambda_i C_a' (1 - e^{-kt})$ in saturation. $Q_{i(\infty)} = V_i \lambda_i C_a'$. For the whole body, or a multiple compartment tissue, $Q = Q_{1(\infty)}(1 - e^{-k_1 t}) + Q_{2(\infty)}(1 - e^{-k_2 t}) + \ldots Q_{n(\infty)}(1 - e^{-k_n t})$ in saturation and similarly for desaturation:

$$Q = Q_{1(o)} e^{-k_1 t} + Q_{2(o)} e^{-k_2 t} + \ldots Q_{n(o)} e^{-k_n t}$$

Most investigators have found at least 2-4 components in their saturation and desaturation curves with ^{85}Kr. The exceptions have been when a GM counter was being used to count the beta emissions from a homogenous surface layer thicker than the maximum beta range such as the cerebral cortex or renal cortex. Wherever NaI detectors were used or heterogenous tissues were counted, monoexponential curves were rarely found (15-32).

Experimental observations in dogs, cats, and humans (4, 6, 7, 10, 15-32) indicate that there are three or four groups of similarly perfused tissues that can be treated as compartments for purposes of whole-body analyses. There is a fast component, with T 1/2 from 0.04-0.8 min, which may be due to arterial blood content. Three slower compartments are usually seen with average half times of about 2.5 minutes, 7.5 minutes, and 20-30 minutes (range of 1-5, 5-10 and 11-180 minutes). Anything that changes blood flow rate can radically change the observed compartmental half-times in saturation or desaturation and can even cause an apparent change in the number of compartments observed.

Lesser and Zak (33) proposed a three-compartment parallel model consisting of:

1. Rapidly perfused lean tissues such as the heart, brain, kidneys, etc.
2. More slowly perfused lean tissues such as resting muscle, skin, connective tissue, etc.
3. Adipose tissue.

While this is obviously an oversimplification, it will suffice for the purpose of this discussion.

The relative contribution of the various compartments to the whole-body burden at full saturation (Q_i) is determined by the size of the compartment, V_i, the tissue:air partition coefficient, λ_i, and the gas concentration in air provided the factors are known. Hypothetical saturation and desaturation curves for standard man are shown in Figure A-1.

Experimental saturation/desaturation data are usually plotted on semilog paper and resolved into components graphically. Computer curve stripping techniques have been reported (34).

Desaturation data obtained with female rats in the experiments previously mentioned were resolved by a stripping technique. The original curve and three components for a rat saturated in an ^{85}Kr mixture for 12 1/2 hours are shown in Figure A-2. The component halftimes were 3.75 minutes, 22.6 minutes, and 94.2 minutes and the corresponding fractions of total activity were 0.248, 0.647 and 0.105. A second experiment, using the same animal and saturation time of 33 1/2 hours, resulted in the two-compartment curves shown in Figure A-3 with halftimes and fractional activities of 15.7 minutes: 0.89, and 85 min: 0.11, respectively. The cause of the differences in curves obtained is not known but may be stress from prolonged exposure to radiation (~ 3000 rad) and chamber dryness resulting in increased respiration and circulation which increased the rate of gas exchange from the tissue to air. Another possibility is greater loss of stored fat in the longer experiment. A greater weight loss was noted; but, since no food or water balance was kept, the loss could not be unequivocally attributed to fat loss.

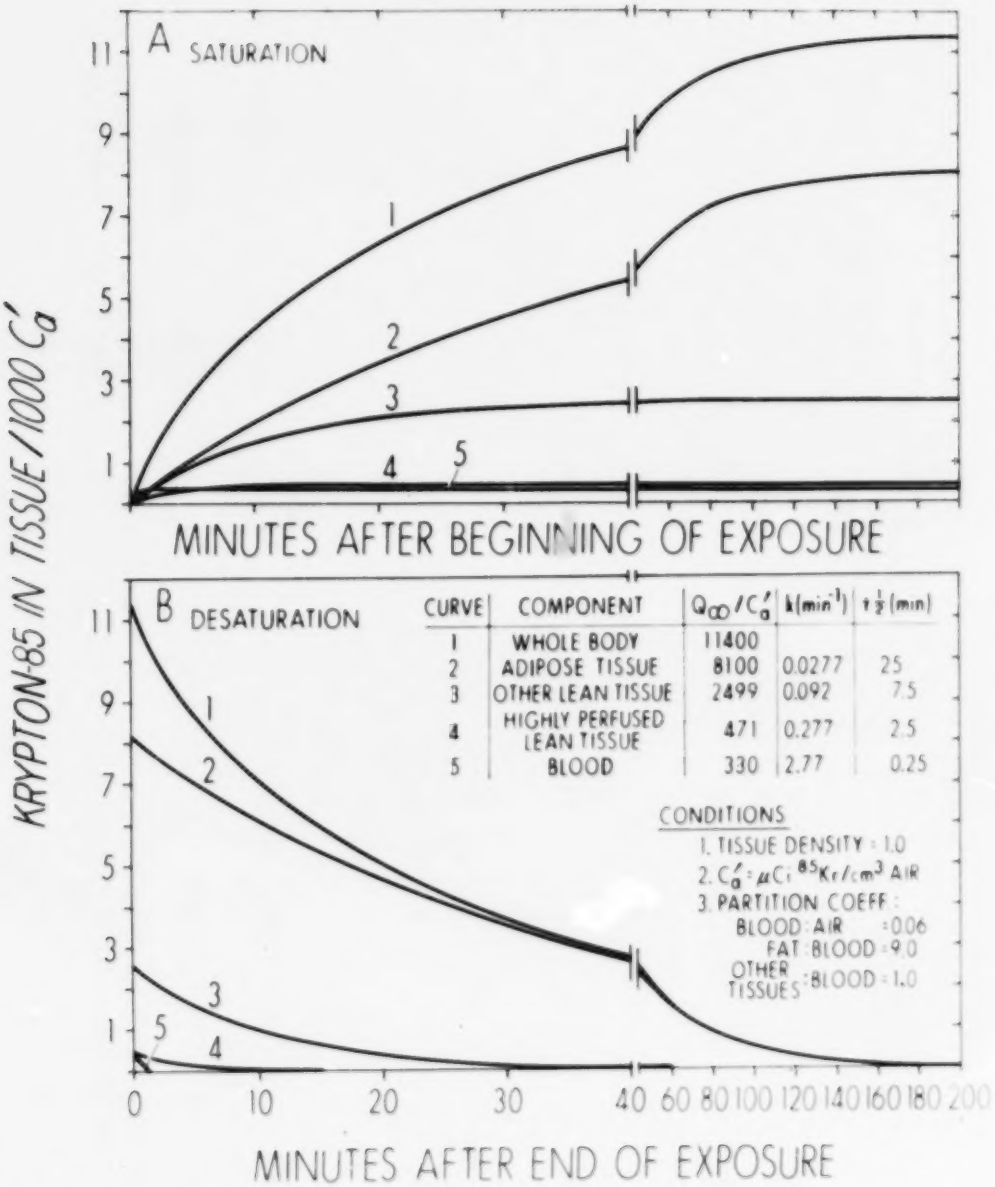
A SATURATION
B DESATURATION
KRYPTON-85 IN TISSUE / 1000 C'a
MINUTES AFTER BEGINNING OF EXPOSURE
MINUTES AFTER END OF EXPOSURE
CURVE | COMPONENT | Q∞/C'a | k(min⁻¹) | t½(min)
1 | WHOLE BODY | 11400 | |
2 | ADIPOSE TISSUE | 8100 | 0.0277 | 25
3 | OTHER LEAN TISSUE | 2499 | 0.092 | 7.5
4 | HIGHLY PERFUSED LEAN TISSUE | 471 | 0.277 | 2.5
5 | BLOOD | 330 | 2.77 | 0.25
CONDITIONS
1. TISSUE DENSITY = 1.0
2. C'a = μCi 85Kr/cm3 AIR
3. PARTITION COEFF:
BLOOD : AIR = 0.06
FAT : BLOOD = 9.0
OTHER TISSUES : BLOOD = 1.0

Figure A-1. Hypothetical ^{85}Kr saturation and desaturation curves for standard man.

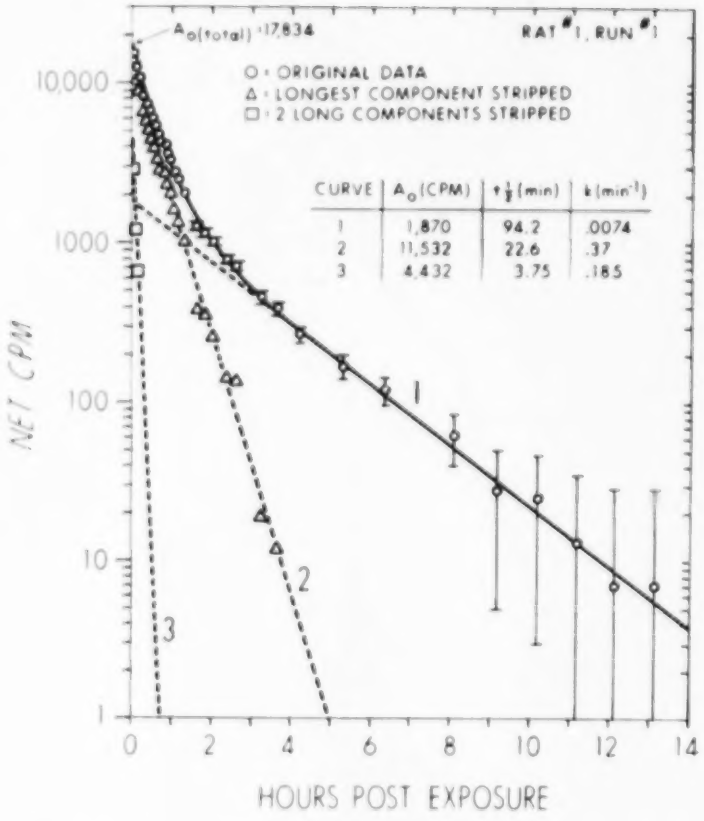
A_o(total) =17,834
RAT # 1, RUN # 1
O = ORIGINAL DATA
Δ = LONGEST COMPONENT STRIPPED
□ = 2 LONG COMPONENTS STRIPPED
CURVE | A_o(CPM) | $t_{\frac{1}{2}}$(min) | k(min⁻¹)
1 | 1,870 | 94.2 | .0074
2 | 11,532 | 22.6 | .37
3 | 4,432 | 3.75 | .185
NET CPM
HOURS POST EXPOSURE

Figure A-2. Experimental ^{85}Kr desaturation curves in rat - short exposure.

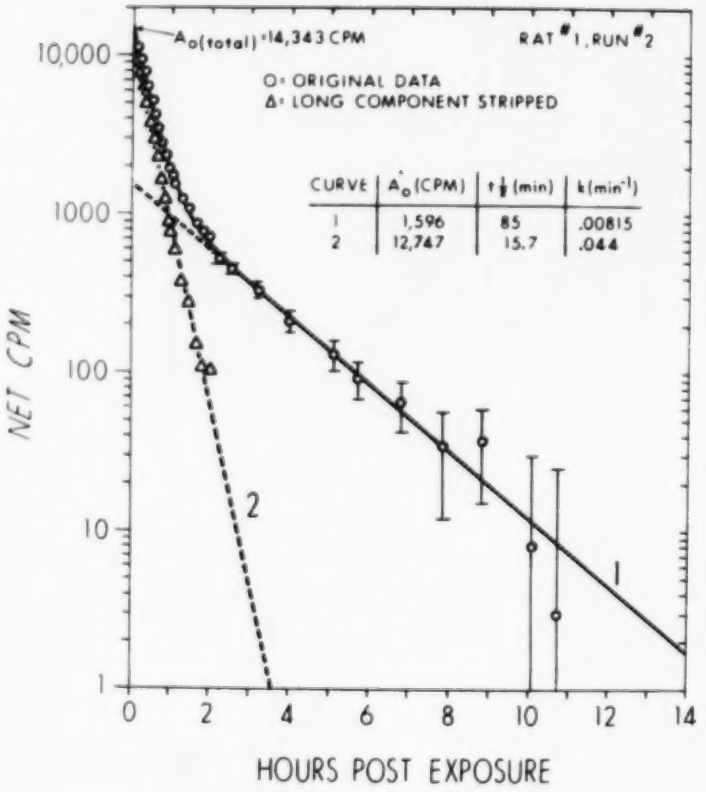
A_o(total) =14,343 CPM
RAT # 1, RUN # 2
O = ORIGINAL DATA
Δ = LONG COMPONENT STRIPPED
CURVE | A_o(CPM) | $t_{\frac{1}{2}}$(min) | k(min⁻¹)
1 | 1,596 | 85 | .00815
2 | 12,747 | 15.7 | .044
NET CPM
HOURS POST EXPOSURE

Figure A-3. Experimental ^{85}Kr desaturation curves in rat - long exposure.

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